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Generalized gamma-ray isotopic analysis software and re-evaluation of the two important 242Pu branching ratios

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Generalized gamma-ray isotopic analysis software and re-evaluation of the two important ^{242}Pu branching ratios*

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Abstract

The goal of the generalized gamma-ray analysis software is to provide precise and accurate isotopic analyses of samples that do not have a standard experimental geometry. This analysis tool will analyze gamma-ray data from all types of measurement scenarios with little or no interaction from the user. This tool will also provide complete transparency regarding the gamma-ray peaks and branching intensities used in the analysis with the capability for the user to modify this information. We are currently at the data collection phase of building a validated spectral library. One of the by-products in this data collection phase is enabling us to reevaluate the two important branching ratios in ^{242}Pu. These branching ratios are required for very high burn-up plutonium fuels. Our preliminary analysis shows that the energy of the states are 103.5 keV and 158.82 keV, the branching ratio are 1.36E-5 (9%) and 3.37E-6 (7%), respectively. More accurate measurements and analysis are currently being carried out.

Introduction

A common shortcoming of high resolution gamma-ray isotopic measurements is the inability to produce good results without user interaction when the intervening material is not well known. We are working to addresses the problem of making accurate isotopic analyses of samples that do not have a standard experimental geometry by developing a more general purpose, gamma-ray radionuclide characterization tool to enable analyses of most gamma-ray spectra for isotopic information. This analysis tool will analyze gamma-ray data from all types of measurement scenarios with minimal interaction from the user. This tool will also provide complete transparency regarding the gamma-ray peaks and branching intensities used in the analysis with the capability for the user to modify this information.

By using a source characterization algorithm in contrast to an identification algorithm, we hope to determine as much information about the source as possible. For example, nuclides identification and intensity, shielding, and the maximum amount of unobserved sources that could be present. To reduce the free parameters in the algorithm, the intrinsic detector efficiency will need to be pre-determined for most of the scenarios.

We are currently at the data collection phase of building a validated spectral library. Three important sources of interest will be measured: plutonium, uranium and fission fragments. After the library collections, we will automate data analysis using various algorithms to determine figure-of-merit of each algorithm. We then choose several methods to ensure the results are unbiased and sufficiently understood. We will prototype and optimize several algorithms to be used as a radionuclide characterization tool, and eventually, write the production code to automate source characterization.

Test library

A few thousand plutonium spectra have been collected to date using HPGe, $LaBr_3$, and CdTe detectors with absorbers ranging from polyethylene to lead and varying thicknesses. Examples of the data are showing in Figure 1, 2, and 3.

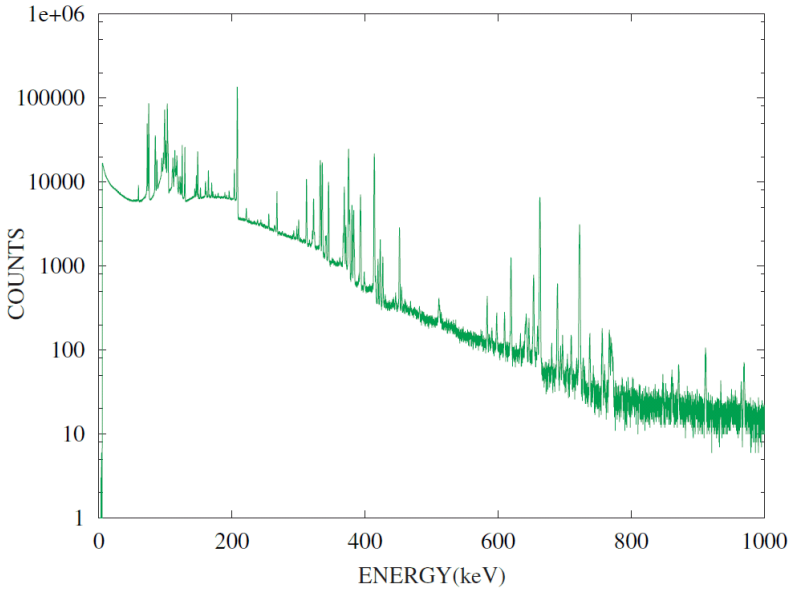

Figure 1. HPGe data of medium burn-up plutonium with 5 mm Fe and 5 in. polyethylene

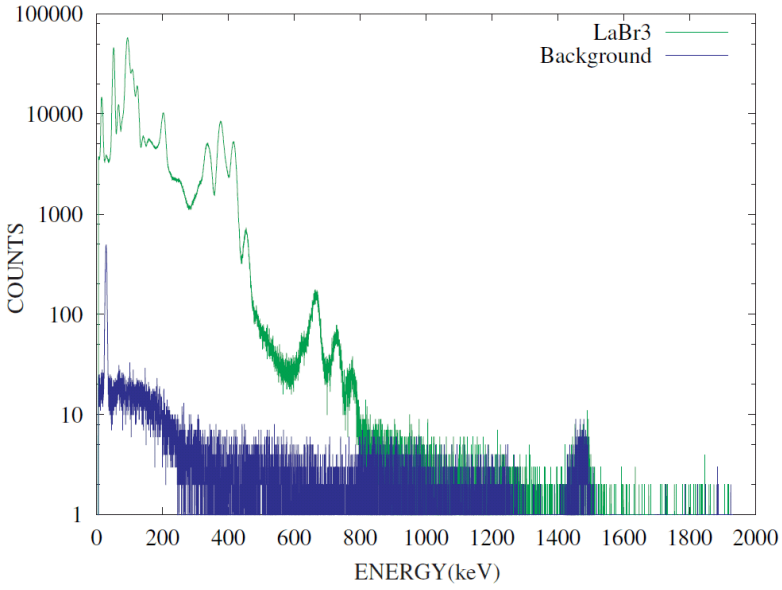

Figure 2 $LaBr_3$ data of a weapon grade plutonium standard with 30 mil cadmium absorber to attenuate the 59-keV ^{241}Am gamma-rays. The intrinsic background of $LaBr_3$ detector is also shown, the ^{138}La electron-capture Kx-rays at 32-38 keV are clearly seen as well as the gamma-ray at 1436-keV. The 789-keV gamma-ray is somewhat hidden in the Compton continuum.

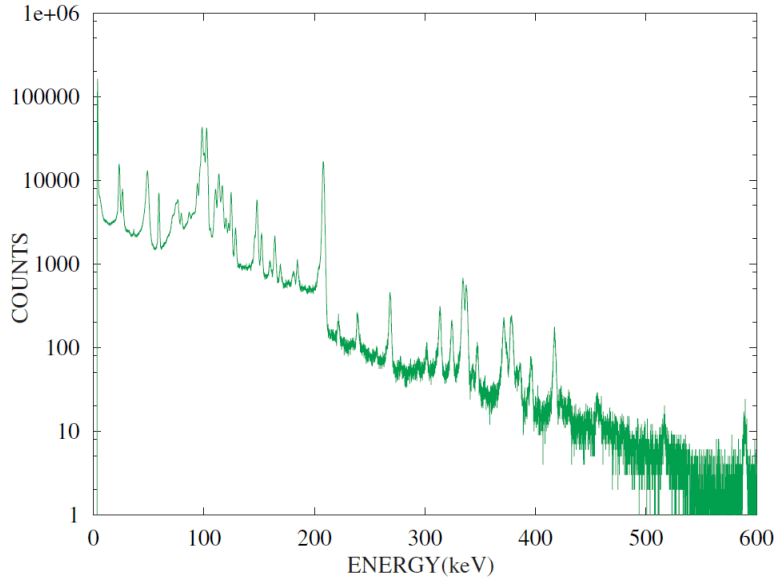

Figure 3 Peltier-cooled CdTe data of high burn-up plutonium with 70 mil of Cd absorber. The energy resolution is better than LaBr3, however, because of the small crystal size, the efficiency above 600-keV is very low.

We are now completing the plutonium spectral library. We are hoping to complete the test suite in the next year.

^{242}Pu problems

When the burn-up is high, the ^{242}Pu isotopic content becomes more important. The traditional correlation method (Ref. 1) will fail. The ^{242}Pu isotopic content in the sample plays an essential role if the neutron coincidence method is used to quantify the total amount of plutonium. During this data collection phase, we had a chance to measure a quite isotopic pure (99.96%) ^{242}Pu sample. We are quite surprised to learn the difference in the branching ratio (BR) value among current nuclear data for the two important gamma-rays at 103.5-keV and 158.8-keV. For example, the Table of Radioactive Isotopes (Ref. 2) values are 7.8(8) E-5 and 4.5(15) E-6, respectively; and the IAEA evaluation (Ref. 3) values are 2.63(9) E-5 and 3.0 (2) E-6, respectively. The LLNL MGA code (Ref. 1) uses the IAEA evaluated value for 103.5-keV and since the MGA code does not use 158.8- keV for ^{242}Pu determination, no information of the branching ratio of this peak is found in the code.

We used a planar detector to collect up to 3 days of data. Figure 4 show the 3 dimensional spectrum overlay onto a weapons grade plutonium spectrum. Because of the age of this standard, the ^{241}Am intensity is quite strong, even though this standard had only 0.0035% of ^{241}Pu twenty nine years ago. A 30 mil Cd absorber was placed between the source and the detectors to attenuate the intensity of the 59-keV ^{241}Am gamma-ray. The total mass of this standard is about 3.5 g.

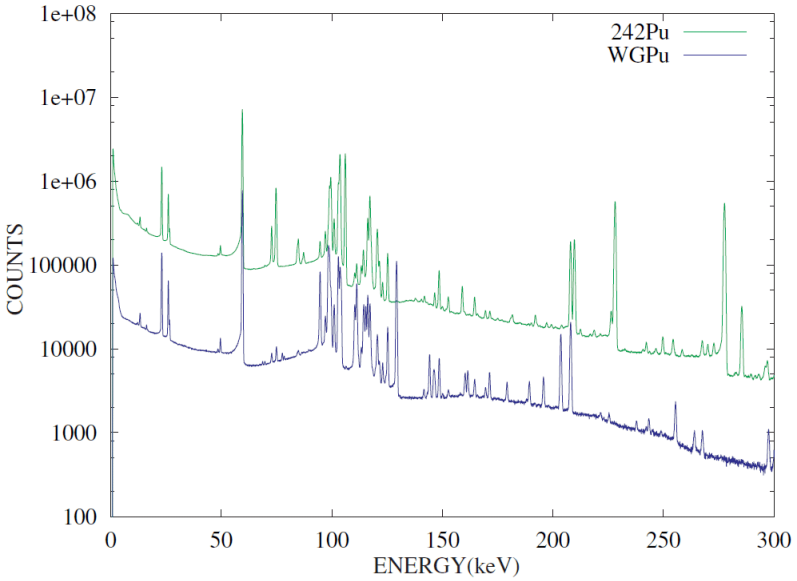

Figure 4. Overlay of the ^{242}Pu spectrum with a weapons grade Pu spectrum

We analyzed the data in two different ways: For the 158-keV peak, we used the nearby ^{241}Pu peak at 160-keV (BR = 6.74E-08) (ref 4) and the ^{241}Pu of this sample (0.0092%) we obtained a branching ratio of the 158-keV, ^{242}Pu peak of 3.37E-06(7%). It is important to note that we used LLNL measured ^{241}Pu BR in the calculation; this number would be 3.29E-6(7%) if the IAEA recommended branching ratio of the 160-keV ^{241}Pu of (6.58E-8) was used. Our result is in good agreement with the IAEA recommended value of 3.0(2)E-06. For the 103.5-keV peak, we used MGA to fit the entire 100-keV region and adjusted the BR of the 103.5-keV to obtain overall agreement with the mass spectrometry data. There are issues observed in the MGA analysis due to low ^{239}Pu content as well as interferences. We were forced to modify the MGA code for this special case. The 100-keV fit of the region in shown in Figure 5. The best BR value of the 103.5-keV is 1.36E-5(9%) which is quite different from values from either the Table of Radioactive Isotopes or the IAEA compilation. We are currently preparing a better measurement to investigate this discrepancy.

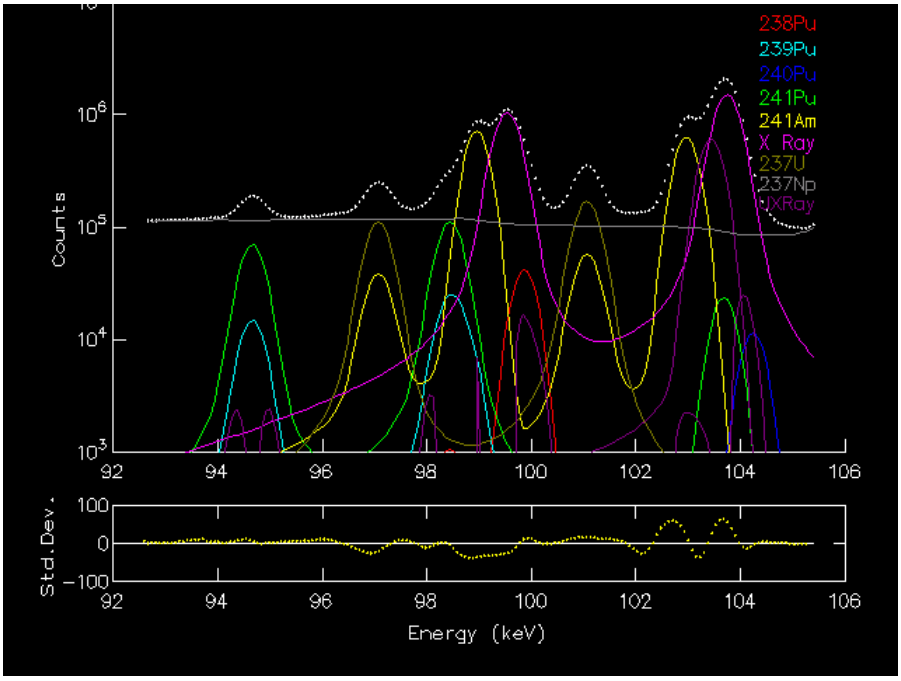

Figure 5. A MGA fit to the 100-keV region of the ^{242}Pu 3-day data. The quality of the fit is poor. This study also reveals some serious issues with current MGA when dealing with weak ^{239}Pu content samples.

Conclusions

To be able to build generalized gamma-ray isotopic analysis code will require a very comprehensive set of high quality spectra using standards. While collecting the data set we had a chance to study the BR issues with the two important gamma-rays in ^{242}Pu. We are in good agreement with IAEA evaluation BR value of the 158.5-keV but not the value of 103.5-keV. We are setting up a better measurement to hopefully obtain an accurate value. The study also revealed some serious issues with the MGA when dealing with "extreme" samples as well as current database issues on the gamma-ray information. The ^{242}Pu study also demonstrated the importance of applying different algorithms to determine figure-of-merit of each algorithm and limitations prior to producing the generalized code.

References

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